Competing anisotropy in the $(Tm_xPr_{1-x})_2Fe_{17}$ system

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Abstract. The magnetization curves of magnetically aligned finely powdered samples of the $(Tm_xPr_{1-x})_2Fe_{17}$ compounds have been measured at 4 K. The easy magnetization axis is oriented in the basal plane or along the hexagonal axis for the compounds with x = 0-0.3 and 0.7-1, respectively. This is because of the absence of magnetic ordering in the Tm and Pr subsystems in these ranges, respectively, and because of competing anisotropy of the subsystems. For the compositions with x = 0.4-0.6, both rare-earth subsystems are magnetically ordered and the easy magnetization axis is oriented between the basal plane and the hexagonal axis. The critical fields of FOMPs decrease quickly as the Pr or Tm content decreases in the ranges 0-0.3 and 0.7-1, respectively. The magnetization anisotropy also diminishes as the Tm content becomes smaller than x = 0.7. No influence of the intrinsic microdeformations on the magnetization of the compounds was detected.

1 Introduction

The Pr_2Fe_{17} and Tm_2Fe_{17} compounds exhibit different crystal structures of the Th_2Zn_{17}- and Th_2Ni_{17}-type and parallel or antiparallel coupling between the moments of the Pr-, Tm- and Fe-sublattices, respectively [1,2]. Pr_2Fe_{17} is a ferromagnet easily magnetized in the basal plane. Tm_2Fe_{17} is unique among the R_2Fe_{17} compounds because it is an easy-axis ferrimagnet below 77 K; at higher temperatures, the magnetization vector lies in the basal plane, and at 234 K, the ferrimagnet transforms to a helimagnet. The temperatures of magnetic ordering equals 284 K for Pr_2Fe_{17} and 274 K for Tm_2Fe_{17}. Thus, a competing anisotropy exists and a concentration spin-reorientation process has to take place in the $(Tm_xPr_{1-x})_2Fe_{17}$ system at low temperatures. Both the Pr_2Fe_{17} and Tm_2Fe_{17} binaries are characterized by FOMP (first order magnetization process) along the c-axis and in the basal plane, respectively. The magnetization anisotropy in Tm_2Fe_{17} takes place at T = 4 K. Microdeformations in the $(Tm_xPr_{1-x})_2Fe_{17}$ alloys with x = 0.6-0.9 have been earlier detected by means of neutron diffraction at room temperature [3]. Feasibly, these microdeformations stipulate a nonmonotonous variation of the temperatures of magnetic phase transitions in the $(Tm_xPr_{1-x})_2Fe_{17}$ system. It is not clear whether these microdeformations influence the magnetization of the compounds or not. Thus, the properties of the Pr_2Fe_{17} and Tm_2Fe_{17} binary compounds are quite different. In the present paper, the magnetization curves of the $(Tm_xPr_{1-x})_2Fe_{17}$ compounds measured along the hexagonal axis and in the basal plane are studied.

2 Experimental details

Polycrystalline ingots of the $(Tm_xPr_{1-x})_2Fe_{17}$ compounds with x = 0, 0.1, 0.2, 0.3, 0.4, 0.5, 0.6, 0.7, 0.75, 0.8, 0.85, 0.9, 1 were prepared by induction melting [3]. The ingots were homogenized at 1293 K for two weeks and then quenched in water. The phase composition of the samples was investigated by the methods of neutron and X-ray diffraction. The compounds investigated crystallize into the hexagonal Th_2Ni_{17}–type and rhombohedral Th_2Zn_{17}-type structures, the fraction of free α-Fe being no larger than 1-3 wt.%. The MPMS technique was used for magnetic measurements in magnetic fields up to 7 T. The magnetization parallel or perpendicular to the c-axis of the magnetically aligned samples was measured at 4 K. To obtain optimal magnetic alignment, the finely powdered samples were magnetically aligned by means of the sample-rotation magnetic alignment technique, as in Ref. [4]. This alignment was performed at the temperatures T=270-272 K which are somewhat lower than the temperatures of the magnetic ordering of all the alloys except for the alloys with x=0.7-0.8 [3]. The X-ray diffraction patterns indicate a good magnetic alignment in the case of the single-phase compounds with x=0-0.3 (rhombohedral structure) and x=0.8-1 (hexagonal structure). A working magnetic alignment was realized in the two-phase compounds with x=0.7, 0.75. The magnetic alignment in the compounds with x = 0.4-0.6 was not successful, as it follows from the X-ray diffraction data.

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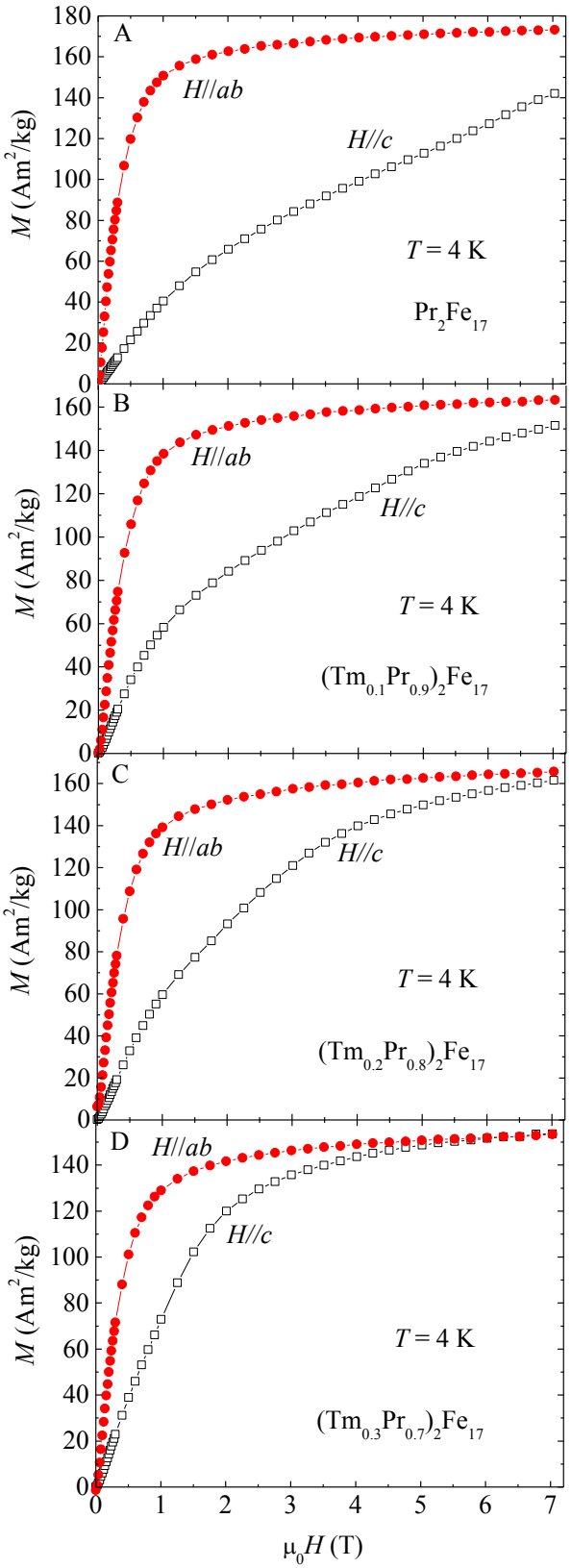

Fig. 1. Magnetization measured at 4 K on magnetically aligned $(Tm_xPr_{1-x})_2Fe_{17}$ in external field applied along the *ab* plane (●) and the *c*-axis (□), *x*=0 (A), 0.1 (B), 0.2 (C), 0.3 (D).

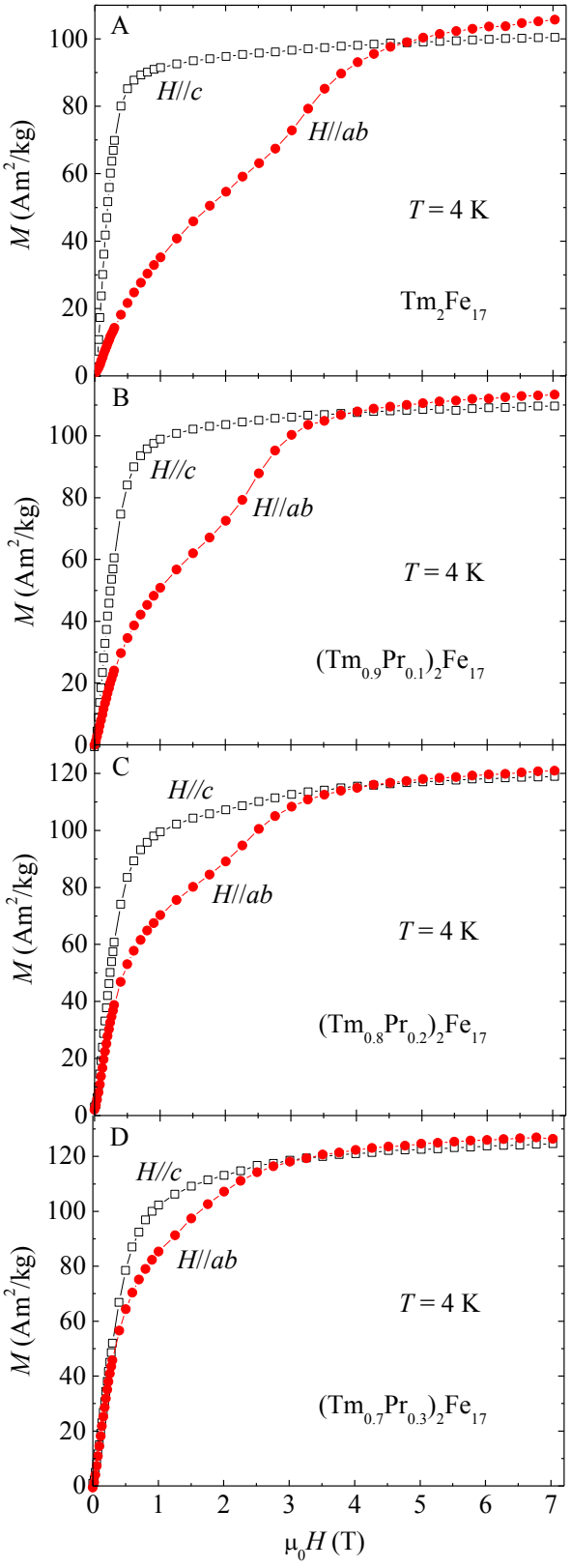

Fig. 2. Magnetization measured at 4 K on magnetically aligned $(Tm_xPr_{1-x})_2Fe_{17}$ in external field applied along the *ab* plane (●) and the *c*-axis (□), *x*=1 (A), 0.9 (B), 0.8 (C), 0.7 (D).

3 Experimental results and discussion

The magnetization curves of the compositions with x=0-0.3 and 0.7-1 are shown in Figs. 1 and 2, respectively. The magnetization curves for the binary compounds are about the same as in Ref. [4]. One can suppose from the magnetization data (Figs. 1, 2) that the magnetic anisotropy in the compositions with x=0-0.3 and 0.7-1 is an easy-plane type or an easy-axis type, respectively, or close to them. Such different orientations of the easy-magnetization axis in the $(Tm_xPr_{1-x})_2Fe_{17}$ system is due to the competing anisotropy of the Tm and Pr subsystems. The FOMP with lower critical fields as compared with the binary compounds is retained for the compositions x=0.1, 0.2, and 0.7-0.9. The magnetization anisotropy of Tm_2Fe_{17} decreases when Pr is substituted for Tm. This result confirms the supposition from Ref. [4] that the magnetization anisotropy of Tm_2Fe_{17} is due, in particular, to canting of the magnetic moments in the Tm-subsystem under the external field applied along the basal plane.

The magnetization curves of the magnetically aligned compositions with x = 0.4-0.6 are about the same along different directions, i.e. these samples are truly polycrystalline. This is in agreement with the data of X-ray diffraction testifying to the absence of magnetic alignment in the samples with x = 0.4-0.6. The non-single-phase state of the compounds with x = 0.5, 0.6 and a harmful effect of magnetic moments of Tm at x = 0.4 (see below for more details) can be the reasons for this unsuccessful alignment. The orientation of the easy-magnetization axis of the compounds with x=0.4-0.6 was not obtained from our magnetization data. Apparently, the easy-axis orientation in the $(Tm_xPr_{1-x})_2Fe_{17}$ compounds with x = 0.4-0.6 changes gradually with the composition variation in accordance with the content of the Th_2Zn_{17}- and Th_2Ni_{17}-type structures. Feasibly, the magnetic neutron diffraction technique will be useful to study the easy-axis orientation in the compositions with x = 0.4-0.6.

Fig. 3 shows the concentration dependences of the magnetization M of the $(Tm_xPr_{1-x})_2Fe_{17}$ compounds measured along the c-axis and in the basal plane in a field of 7 T at T = 4 K. The M values along these two different directions are close to each other for x = 0.2-1. The noticeable difference appears only for the compositions with x=0 and 0.1 because of their large magnetic anisotropy (Fig. 1), as is for the Pr_2Fe_{17} compound in Ref. [4]. As a result, the magnetization along the basal plane decreases monotonically with increasing x, whereas the magnetization along the c axis does not change monotonically in the range x = 0-0.3. We suppose that the spontaneous magnetic moments of the Pr and Fe subsystems lie in the basal plane for the compounds with x = 0-0.3. The Tm subsystem is not magnetically ordered in the compounds with x = 0.1-0.3, as one can suppose from the data on Fig. 1. The Tm magnetic moment has to be oriented against the magnetic moments of the Fe and Pr ions in the basal plane, whereas the easy-axis type magnetic anisotropy of the Tm ions causes the orientation of the Tm moments along the c axis. Apparently, the competition of these two interactions causes the spontaneous orientation of the Tm moments at an angle to the c-axis for x = 0.1-0.3, and the Tm moments create a fan around the c-axis. The magnetic field applied along the c-axis deflects the moments of the Pr and Fe ions from the basal plane and change the angle of the fan of the Tm moments. This picture allows us to explain the non-monotonic variation of the magnetization along the c-axis in a field of 7 T (Fig. 3). The decrease in the contribution from the Pr subsystem to the total magnetization is compensated for or even exceeded by the increase in the contribution from the Tm subsystem for the compositions with x = 0.1-0.3.

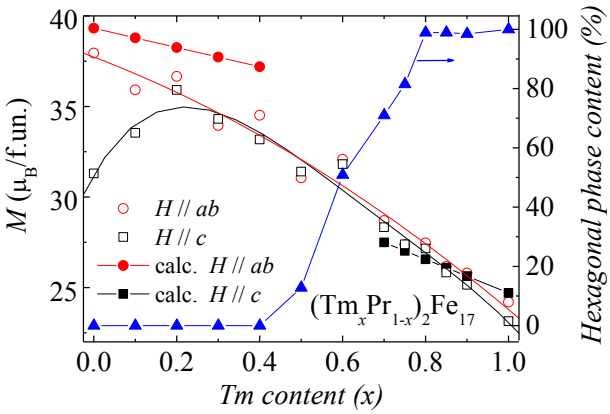

Fig. 3. Concentration dependences of the magnetization M of the $(Tm_xPr_{1-x})_2Fe_{17}$ compounds measured along the ab plane (o) and the c-axis (□) at T = 4 K in a field of 7 T. The M values for x = 0.4-0.6 were measured for the polycrystalline samples (see text). The estimated M values along the ab plane (●) and the c-axis (■) are presented, see text for details. The percentage content of the Th_2Ni_{17}-type hexagonal phase is shown by (▲) [3].

Let us estimate the concentration variation of the magnetization of the $(Tm_xPr_{1-x})_2Fe_{17}$ compounds measured in a field of 7 T applied along the c axis and in the basal plane. We suppose that the spontaneous magnetization lies in the basal plane or along the c axis (or close to these crystallographic directions) for the compounds with x=0-0.3 and 0.7-1, respectively. Such supposition can be made from the $M(H)$ curves in Figs. 1 and 2. We take into account a real composition, $Tm_2Fe_{19.35}$, for the compounds with x=0.7-1, as it was obtained by means of neutron diffraction technique in Ref. [3]. The magnetic moments of the Pr, Tm and Fe ions were taken as 2.66 μ_B [1], 7 μ_B [2] and 2 μ_B, respectively. As can be seen from Fig. 3, there is a good fit of the estimated data to the experimental ones for the compounds with x = 0.7-1 measured along the c axis (along the basal plane in the case of x=1 because of the strong magnetization anisotropy). The estimated data of the magnetization measured along the basal plane in the compounds with x = 0-0.3 are larger than the experimental ones, perhaps, because of the neglect of the magnetic anisotropy of the Pr ions in the basal plane and contribution from the Tm sublattice. The large difference

between the calculated and the experimental data for x=0.4 can be explained by the orientation of the easy magnetization axis of the compound at an angle to the basal plane. This is an interesting result because the composition with x=0.4 crystallizes in the rhombohedral phase with the intrinsic easy-plane type magnetic anisotropy. The result can be explained by the contribution of the easy-axis type anisotropy of the Tm ions, which substitute in part for the Pr ions in the rhombohedral structure and have a much larger magnetic moment.

Microdeformations do not influence the magnetization of these alloys (as one can see from Fig. 3) unlike the early obtained strong decrease of the temperatures of the magnetic phase transitions for the compositions with x = 0.6-0.9 [3]. This result confirms the supposition from Ref. [3] that an inhomogeneous distribution of Pr and Tm atoms in the defected hexagonal lattice and the large difference of their atomic radiuses can be the reasons for the nonmonotonic variation of the temperatures of the magnetic phase transitions in the system. It is well known that the temperatures of the magnetic phase transitions in the R_2Fe_{17} compounds strongly depend on the interatomic Fe-Fe distances [1-3].

4 Conclusions

It has been established that the easy magnetization axis in the $(Tm_xPr_{1-x})_2Fe_{17}$ compounds is oriented in the basal plane for x = 0-0.3 or along the hexagonal axis for x = 0.7-1 at T = 4 K. This is because of the absence of magnetic ordering in the Tm and Pr subsystems in these ranges, respectively, and because of competing anisotropy of the subsystems. For the compositions with x = 0.4-0.6, both these rare-earth subsystems are magnetically ordered and the easy magnetization axis has to be oriented between the basal plane and the hexagonal axis. To study the easy axis orientation in the compositions with x = 0.4-0.6 it is necessary to use the magnetic neutron diffraction technique. The critical fields of FOMPs decrease quickly as the Pr or Tm content decreases in the ranges 0-0.3 and 0.7-1, respectively. The magnetization anisotropy also diminishes as the Tm content becomes smaller x = 0.7. No influence of the intrinsic microdeformations on the magnetization of the compounds was detected unlike the early obtained strong decrease of the temperatures of the magnetic phase transitions for the compositions with x = 0.6-0.9 under the action of these microdeformations [3]. This result confirms the supposition from Ref. [3] that an inhomogeneous distribution of Pr and Tm atoms in the defected hexagonal lattice and the large difference of their atomic radiuses can be the reasons for the nonmonotonic variation of the temperatures of the magnetic phase transitions in the system.

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